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JAN 81 B CRASEMANN, M H CHEN

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) X-Ray and Auger transition probabilities were calculated for atoms that have been singly and multiply ionized in inner shells. The effects of relativity and configuration interaction on these rates were studied, and lifetimes of atomic inner-shell holes were computed. Synchrotron radiation was employed for precision measurements of atomic level energies, for the study of inelastic scattering, and for the investigation of sub-threshold excitation of atomic hole states.		

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ATOMIC INNER-SHELL PROCESSES.

Final Report

B Bernd/Crasemann
Mau Hsiung/Chen

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1. STATEMENT OF PROBLEMS STUDIED

X-ray and Auger transition probabilities were calculated for atoms that have been singly and multiply ionized in inner shells. The effects of relativity and configuration interaction on these rates were studied, and lifetimes of atomic inner-shell holes were computed. Synchrotron radiation was employed for precision measurements of atomic level energies, for the study of inelastic scattering, and for the investigation of sub-threshold excitation of atomic hole states.

2. SUMMARY OF MOST IMPORTANT RESULTS

A central topic of our research has been the study of a perplexing systematic discrepancy between theoretical and measured atomic inner-shell level widths or hole-state lifetimes. This discrepancy is most striking (a factor of 2 or 3) for 2s vacancy states specifically, and for states deexcited by low-energy Coster-Kronig transitions in general. The discrepancy is now essentially understood; it is explained in terms of the following factors: (i) exchange and relaxation effects play a major role in low-energy Coster-Kronig transitions, (ii) relativistic effects can be substantial, even for medium-heavy atoms, and (iii) electron-electron Coulomb correlations can substantially alter the transition rates.

Calculations of multiplet x-ray and Auger rates to $L_{2,3}$ vacancy states of multiply ionized S and Cl, treating the initial state in intermediate coupling, have shown that the spin-orbit interaction has a remarkable effect on the Auger rates and x-ray fluorescence yields of these systems. Clearly,

the correct coupling scheme is of crucial importance in calculations of the lifetimes of inner-shell vacancies; errors of an order of magnitude can otherwise occur.

A general computer code for the relativistic calculation of Auger rates was completed after several years of development. Extensive, systematic Dirac-Hartree-Slater (DHS) computations of Auger rates, level widths, and fluorescence yields have been performed for the K, L, and M shells of atoms throughout the periodic table. Interesting conclusions can be drawn regarding the importance of relativity in the deexcitation of atomic inner-shell vacancies, and regarding the mechanisms through which relativity affects radiationless transition rates.

We used the Møller operator to express (in the local approximation) the relativistic interaction-between two atomic electrons. We have studied the separate effects of (i) relativistic wave functions, (ii) the current-current interaction, and (iii) retardation. A most interesting conclusion is that the effect of relativistic wave functions, i.e., of the Dirac distribution of atomic charge, can profoundly alter transition rates even in outer shells, where the active electrons themselves move at definitely nonrelativistic speeds. This consequence arises because the inner electrons are drawn closer to the nucleus in the DHS potential, causing more screening, so that the outermost electrons move out further from the nucleus than in the nonrelativistic approximation. The intensity of weak transitions in the M shell, for example, is consequently observed to change by as much as a factor of two when relativity is included.

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In inner shells, not only relativistic wave functions, but also current-current interaction and retardation can greatly affect the radiationless decay rates. For $K-L_1L_1$ transitions, for example, each of these effects approximately doubles the rate at $Z=70$. For some other classes of transitions (e.g. $K-L_3L_3$), the rates are reduced by comparable factors when relativity is included. For some rates (e.g., $K-L_2L_3$), the current-current interaction has the opposite effect from the retardation, so that the net effect of relativity is slight. Cancellations of this kind also tend to reduce the effect of relativity on total widths, which are determined by a large number of transition rates. Nevertheless, the total radiationless K-level width is enhanced by relativity, by a factor of ~ 3 at $Z \approx 90$. K-shell fluorescence yields are reduced, and our ab initio calculated K fluorescence yields agree extremely well with the most reliable measurements.

An interesting effect observed in the M shell is that weak radiationless transitions are affected much more by relativity (as much as 100%) than strong transitions. This phenomenon can be understood by noting that weak transitions often involve outer electrons, for which wave-function effects are more pronounced; other weak transitions have small matrix elements because of accidental cancellations that are removed if the potential is altered by going from a nonrelativistic charge distribution to a relativistic distribution; in a third class of weak transitions, the two final holes are in very different states, and their small wave-function overlap is much more sensitive to the inclusion of relativity than the large overlap of holes in similar states.

In summary, we find: (1) The effect of relativity on Auger rates, level widths, and fluorescence yields is diverse and can be very pronounced even

for outer levels; (2) the mechanisms through which relativity affects radiationless transition rates can be understood in terms of the separate effects of relativistic wave functions, current-current interaction, and retardation; (3) our relativistic calculations of the lifetimes of atomic inner-shell vacancies, level widths, and fluorescence yields are in far better agreement with the (generally scant) experimental data than previous, nonrelativistic theoretical results; (4) our ab initio calculations of Auger spectra agree well with measured spectra if one includes (i) relativity, (ii) intermediate coupling where appropriate, and (iii) configuration interaction.

In our experimental program at the Stanford Synchrotron Radiation Laboratory, we developed a new method to determine inner-shell level energies of noble gases with very high accuracy. Absorption edges were measured at high resolution, using synchrotron radiation monochromatized with a channel-cut crystal that was calibrated through a novel procedure. The absorption edges were modelled theoretically, and the theoretical edge shapes were fitted to the measured spectra. The results have been compared critically with ab initio theoretical energies, leading to tests of quantum-electrodynamic shifts and relativistic corrections in the theory.

In a collaboration with investigators at the Institut du Radium in Paris, we have made a theoretical comparison of electron excitation probabilities accompanying (1) orbital electron capture by the nucleus and (2) photoionization or ionization by electron impact. Not only is the excitation probability during nuclear electron capture much lower than with other primary ionization mechanisms, but it also preferentially selects different subshells. The calculations

explain x-ray satellite observations first made by the Paris group. In more general terms, this study draws attention to the need for much further experimental and theoretical work on shakeup and shakeoff processes, that truly epitomize electron correlation effects.

Probably the most important outcome of our experimental program is the discovery of the Auger resonant Raman effect. This was the first major result obtained with our on-line gas-phase electron spectrometer installed in the Stanford Synchrotron Radiation Laboratory. In this experiment, we used monochromatized hard synchrotron radiation near the photoionization threshold to produce the $2p_{3/2}^{-1}$ vacancy state in atomic Xe. Deexcitation of the state through $L_3-M_4M_5(^1G_4)$ Auger-electron emission was measured. The 5d spectator-electron Auger satellite was observed. The satellite energy was found to exhibit linear dispersion as a function of the photon energy of the exciting radiation. The observed width of the 1G diagram line was found to decrease by -40% at threshold. This new radiationless process could thus be identified as the Auger analog of the x-ray resonant Raman effect. Post-collision interaction was seen to shift the diagram line by -3 eV, or approximately one thousand time more than previously observed post-collision interaction. We expect to perform extensive additional work on sub-threshold excitation of inner-shell vacancy states, which is a new field in atomic physics that calls for theoretical as well as experimental exploration.

3. PUBLICATIONS

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4. SCIENTIFIC PERSONNEL

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